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| 14. ABSTRACT We have four main results from the research supported by the AFOSR over the last three years. They are (1) a new mode of Stark deceleration to increase the energy resolution of the resulting molecular beam, (2) theoretical investigation of resonant collisional quenching of NH molecules, (3) measurement of electric field affected collisions between cold ammonia molecules and ultracold rubidium atoms, (4) creation and deceleration of velocity controlled beams of rotationally excited ammonia molecules. | | | | | | |
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High-energy-resolution molecular beams for cold collision studies

The first goal of the project was to develop a new slowing method for a Stark decelerator to decrease the energy spread in the slowed molecular packet and increase the temporal and spatial separation of the slowed packet from an unsloved beam. This new method would increase the usefulness of such energy-tuned beams that could be used for studying collisions with narrow resonances and thresholds. We published an article on these results in a special issue of the New Journal of Physics, which was dedicated to cold and ultracold molecules. Fig. 2 summarizes the main results of this work.

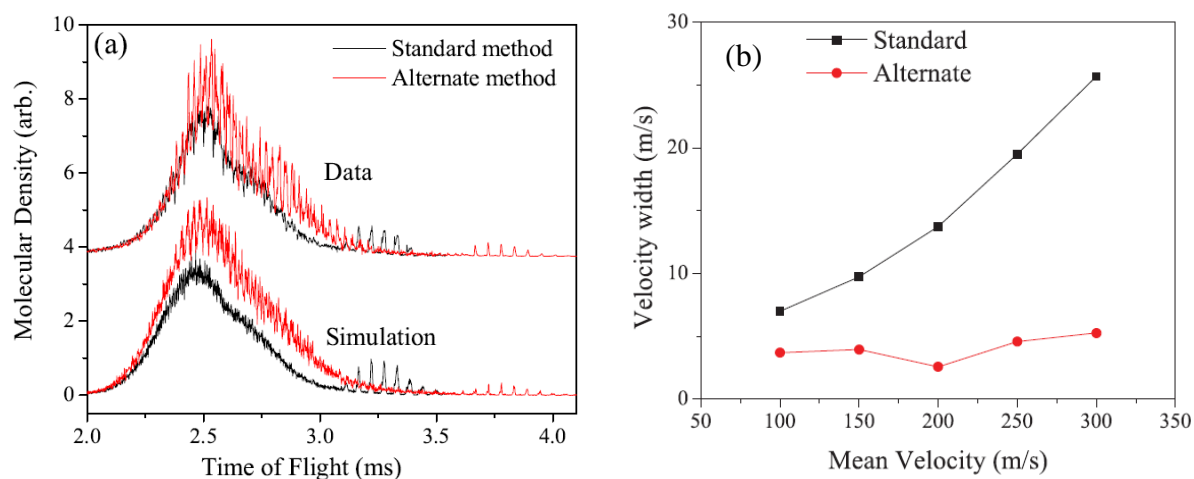


Figure 2. Results from the alternate slowing protocol (a) Time-of-flight traces showing the arrival time of decelerated peaks using both standard and alternate methods of slowing. With the alternate deceleration method, the decelerated peak arrives 515 μ s later than the analogous peak using the standard method. The results from a 3D Monte-Carlo simulation are also shown (lower curves). (b) The velocity widths of molecular packets for the standard and alternate deceleration methods. For standard slowing, there is a strong correlation between the final velocity and the velocity width of the decelerated packet. In the alternate slowing method, however, the velocity width is effectively constant as a function of final velocity.

Resonant Collisional Quenching

There is currently a lot of interest in molecule interactions where the molecule is in a single electronically or rotationally excited state. There are many processes that involve excited-state molecules including chemical reactions, photodissociation, and collisions. One active area of research involves studying subsequent processes after excitation. There are many fundamental questions to be answered such as: How does the energy flow within a molecule or get transferred to another molecule? How does the presence of the excitation energy change molecular interactions and chemical reactions? In addition, systems that have collisional resonances are becoming increasingly

important as control over the reactants becomes increasingly precise. We choose to study NH as it has a nearly identical low-level energy structure to O₂, whose metastable state plays a large role in the dynamics of the upper atmosphere.

In collaboration with Chris Greene's theory group, we examined the quenching reaction $\text{Rb } (^2\text{S}) + \text{NH } (^1\Delta) \rightarrow \text{Rb } (^2\text{P}_{1/2}) + \text{NH } (\text{X } ^3\Sigma^-) \rightarrow \text{Rb } (^2\text{S}_{1/2}) + \text{NH } (\text{X } ^3\Sigma^-) + h\nu$. One can think of this channel as converting the energy stored in the doubly forbidden metastable NH molecule to a *photon*. This reaction is interesting in that it involves initial and final states that are nearly degenerate. This near degeneracy is expected to lead to a large reaction rate. We examined this system using *ab initio* quantum chemistry calculations and scattering calculations, which included spin-orbit effects. We found that the reaction rate is large and, in fact, approaches the quantum mechanical unitarity limit. This work was published in Physical Review A. Now that we have an estimate of the effect of the resonance, we are about to experimentally measure the dynamics of the NH+Rb system. This will be one of the first experiments conducted in the new traveling-wave decelerator.

Electric field affected cold collisions between molecules and atoms

One of the tantalizing aspects of creating systems of cold molecules is the possibility of controlling the microscopic interactions with external electric and magnetic fields. Controlling the interactions could lead to a better understanding of the role of quantum mechanics in molecular collisions and in the fundamental processes of breaking and forming a chemical bond. There have been considerable theoretical investigations of how long-range dipole-dipole interactions between two polar molecules can be affected by the application of an electric field to control the relative orientation of the molecules. However, there has been only one system that has reached the densities of polar molecules required to study these effects. We show that even in the absence of the electric-dipole interaction the space-orienting nature of electric fields can change collision dynamics. We measure elastic and inelastic collision cross-sections between rubidium atoms (Rb) and ammonia

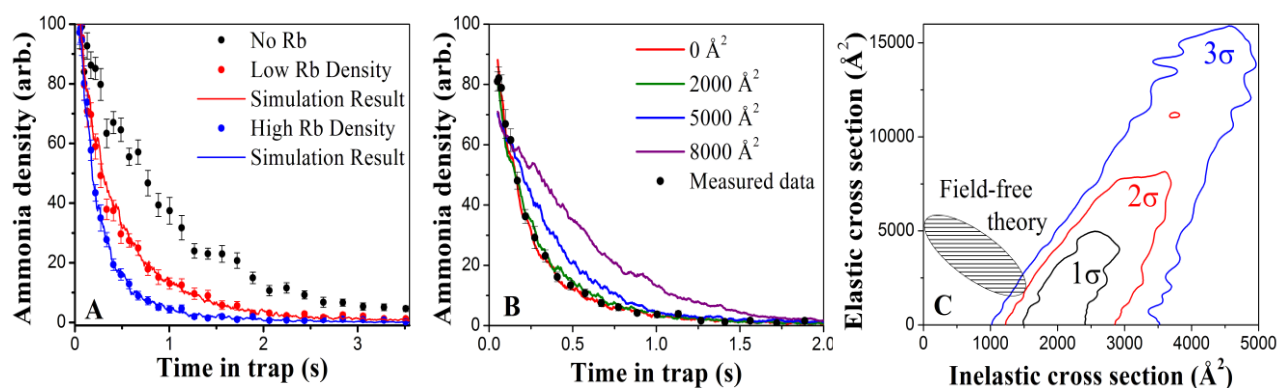


Fig. 1: Measured decay curves and simulation results for ND₃ and extracted elastic and inelastic cross sections. (A) The peak ammonia density is plotted as a function of time for various densities of Rb, including the corresponding best-fit simulations. (B) The effect of elastic collisions on the simulated ND₃ decay profile. The density decay at the trap center is reduced for larger elastic cross sections as a result of ND₃ migrating towards the trap center due to sympathetic cooling with Rb. (C) Confidence contours for the elastic and inelastic cross sections extracted from simulations of the experimental decay curves. The shaded oval represents the range of possible results from field-free quantum-mechanical scattering calculations and is obtained by repeating the calculations for a variety of scaled potential-energy surfaces.

molecules (ND_3) in the presence of an electric field in the cold temperature regime (~ 100 mK).

We use a co-trapped geometry for these experiments where the molecules are decelerated and subsequently trapped using static electric fields and the rubidium atoms are laser cooled and then trapped using magnetic fields. Once each species is trapped in separate regions of the vacuum system, the coils forming the magnetic trap are translated 60 cm to overlap the two clouds initiating the interactions. We then monitor the dynamics of the ND_3 density by measuring the number of ions created via resonance enhanced multi-photon ionization. We can also measure the temperature and density of the Rb using resonant absorption imaging. Using these measurements and associated Monte Carlo simulations to model some of the dynamics, we can extract the inelastic collision cross-sections and place upper limits on the elastic cross-section (Fig. 1).

The inelastic cross-sections are nearly at the Langvin capture limit and factors of 5-10 above initial theoretical calculations done by the J. Hutson group at Durham, UK. However, the initial calculations were done without an electric field present. We have worked with the Hutson group and they have now included the effect of electric fields. We see an increase in the inelastic cross-sections with increasing electric fields.

One can understand this increase using a semi-classical picture of the atom-molecule interactions. Based on the calculated potential surface, the atom always wants to approach the molecule from the nitrogen end. At low electric fields, ND_3 will adiabatically rotate such that it hits nitrogen end first and then comes back out on the same adiabat with its internal state unchanged. For incredibly large electric fields, the ammonia molecule is essentially fixed in space and the Rb atom will collide at a random angle. For intermediate electric fields, there will be a distance where the energy of the Stark shift becomes comparable to the energy to rotate the molecule. There is now a frustration between the orientation due to the electric field and the orientation due to the incoming Rb atom. In this situation, an avoided crossing between two different states in ammonia appears and thus causes an increased probability of the molecule changing state during the collision. Thus, even without dipole-dipole interactions, externally applied electric fields can change the microscopic collision dynamics. This work was published in Phys. Rev. Lett.

Stark Deceleration of Rotationally Excited Molecules

Over the last decade, several new methods have been developed to create cold trapped samples of ground state molecules including Stark deceleration, which uses inhomogeneous electric fields to decelerate a molecular beam. These trapped samples allow for investigations of chemical reactions where the collision energy can be reduced to the 1-100 mK range.

We have increased the level of control over the external motion of molecules, but creating a molecular beam with just one internal state populated is also important. Experiments that explore scattering on a state-by-state level at cold temperatures can reveal detailed information about molecular interactions. In recent years, Stark deceleration has emerged as one of the main methods for producing velocity-controlled molecular beams. It is widely assumed that Stark deceleration is also highly quantum-state selective as the dynamics of the deceleration process depend heavily on the Stark shift and hence on the molecular quantum state. However, we show that Stark deceleration cannot effectively produce a molecular beam consisting of a single quantum state when other weak-field-seeking states are present in the initial beam. Therefore, quantum state purity must be carefully considered when using Stark decelerated beams, particularly in collision experiments where contributions from all rotational states must be addressed. We have manuscript under review at New Journal of Physics describing these measurements and the implications of the results